arXiv:0712.2250v1 [cond-mat.str-el] 13 Dec 2007

# Bose-Einstein Condensation in Magnetic Insulators

Thierry Giamarchi,[1] Christian Rüegg,[2] and Oleg Tchernyshyov[3]

[1]*DPMC-MaNEP, University of Geneva, 24 Quai Ernest Ansermet, 1211 Geneva 4, Switzerland*

[2]*London Centre for Nanotechnology and Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK*

[3]*Department of Physics and Astronomy, Johns Hopkins University, Baltimore,MD 21218, USA*

**The elementary excitations in antiferromagnets are magnons, quasiparticles with integer spin and Bose statistics. In an experiment their density is controlled efficiently by an applied magnetic field and can be made finite to cause the formation of a Bose-Einstein condensate (BEC). Studies of magnon condensation in a growing number of magnetic materials provide a unique window into an exciting world of quantum phase transitions (QPT) and exotic quantum states.**

## INTRODUCTION

Not long after Bose and Einstein described quantum statistics of photons [1] and atoms [2], Bloch applied the same concept to excitations in solids. He treated misaligned spins in a ferromagnet as magnons, particles with integer spin and bosonic statistics, to describe the reduction of spontaneous magnetisation by thermal fluctuations [3]. Matsubara and Matsuda pointed out an exact correspondence between a quantum antiferromagnet and a lattice Bose gas [4]. It is thus natural to ask: can these bosons undergo Bose-Einstein condensation and become superfluid? The answer is yes. The question of order in spin systems and possibility of BEC of magnons has been investigated theoretically for a number of quantum antiferromagnets [5, 6, 7, 8, 9, 10, 11, 12, 13]. Although in simple spin systems the concept of BEC can be applied, in practice factors such as the large value of exchange constants and the presence of anisotropies violating rotational symmetry may restrict their usefulness. However, the analogy between spins and bosons has shown to be very fruitful in those antiferromagnets where closely spaced pairs of spins $S = 1/2$ form dimers with a spin-singlet ($S = 0$) ground state and triplet ($S = 1$) bosonic excitations called variously magnons or triplons. In such systems BEC has been predicted to occur [7] and was experimentally observed [8, 14] in the dimer system $TlCuCl_3$. This started a flourish of activity on the subject and the hunt for such transitions in other magnetic materials.

Other examples of condensates from elementary excitations were recently found in magnetic thin films [15] and semiconductor microcavities [16]. Lattice bosons and several transitions occurring in

these systems have been studied extensively on ultra-cold atomic gases in optical lattices [17, 18, 19, 20]. Here we present an overview of recent developments in the field, and discuss how quantum antiferromagnets offer several advantages that set them apart from the other model systems in which the phenomenon of BEC occurs. We also discuss ways to go beyond the physics of simple BEC condensation and to look at the fascinating new quantum phases of interacting bosons on a lattice.

## BOSONS IN MAGNETS

Let us illustrate the basics of magnon BEC in real dimerized antiferromagnets, such as extensively studied $TlCuCl_3$ [8, 11, 12, 14, 21, 22, 23, 24, 25, 26, 27, 28, 29] and $BaCuSi_2O_6$ [30, 31, 32, 33, 34, 35, 36]. The lattice of magnetic ions in such materials (Fig. 1a) can be visualized as a set of dimers, pairs of copper ions $Cu^{2+}$ carrying $S = 1/2$ each and interacting via Heisenberg exchange:

$$\mathbf{H} = \sum_i J_0\,\mathbf{S}_{1,i}\cdot\mathbf{S}_{2,i} + \sum_{\langle mnij\rangle} J_{mnij}\,\mathbf{S}_{m,i}\cdot\mathbf{S}_{n,j} - g\mu_B H \sum_{\langle ni\rangle} S^z_{m,i}, \tag{1}$$

where $H$ denotes an external magnetic field in $z$–direction, $i$,$j$ number dimers, and, $m, n = 1, 2$ their magnetic sites. The *intra*-dimer exchange is the strongest interaction, which happens to be antiferromagnetic, $J_0 > 0$, so that an isolated dimer has a ground state with total spin $S = 0$ and a triply degenerate excited state of energy $J_0$ and spin $S = 1$ (Fig. 1c). It is convenient to identify the triplet state with the presence of a triplon, a bosonic particle with $S = 1$, and the singlet state with the absence of a triplon, see Fig. 1b. As long as *inter*-dimer interactions are weak, the ground state consists of non-magnetic singlets. It is disordered down to absolute zero temperature without long-range magnetic order. The triplon excitations are made mobile by effective and weak *inter*-dimer couplings $J_{1,2,...} > 0$, from a sum over single–ion interactions $J_{mnij}$, see Fig. 1b. For dimers forming a square lattice (for simplicity) the energy of a triplon with spin projection $S^z = 0, \pm 1$ is

$$\varepsilon(\mathbf{k}) = J_0 + J_1[\cos(k_x a) + \cos(k_y a)] - g\mu_B H S^z, \tag{2}$$

where $\mathbf{k} = (k_x, k_y)$ is the particle wavevector, $a$ is the lattice constant, and $D = 4J_1$ the bandwidth, see Fig. 1c. The energy-momentum dependence of the triplons and singlet-triplet correlations have been measured directly by inelastic neutron scattering [21, 30, 31, 37].

The Zeeman term $-g\mu_B H S^z$ controls the density of triplons. As the magnetic field increases, the excitation energy of triplons with $S^z = +1$ is lowered and eventually crosses zero, as shown in

Fig. 1c and 2a. This defines two critical magnetic fields $H_{c1}$ and $H_{c2}$ in the phase diagram, see Fig. 1d. At zero temperature, below $H_{c1}$ the magnetisation $m_z(H)$ is zero and only singlets exist. Between $H_{c1}$ and $H_{c2}$ the magnetisation increases as the triplon band fills up, see Fig. 1c. Above $H_{c2}$ each site is occupied by a triplon and the magnetisation saturates at one per dimer.

The bosonic nature of triplons is guaranteed by the simple fact that spin operators of two different dimers commute. However, because a dimer can hold at most one triplon, the bosonic picture requires the introduction of a hard-core constraint to exclude states with more than one quasi–particle per dimer. The constraint, which can be interpreted as a strong short-range repulsion between the bosons, poses a difficult theoretical problem. But close to $H_{c1}$ their density is small and collisions between bosons are rare; in this limit interactions can be fully taken into account. By particle-hole symmetry, a similar simplification occurs near $H_{c2}$.

## BEC OF TRIPLONS

The nature of the ground state above $H_{c1}$, its interpretation as a BEC of magnetic quasi-particles, and the tuning of the particle density become particularly accessible if the spin Hamiltonian in Equ. (1) is rewritten in the second-quantized form [7, 8]

$$\mathbf{H} = \sum_i (J_0 - h) a_i^\dagger a_i + \sum_{i,j} t_{ij} a_i^\dagger a_j + \frac{1}{2} \sum_{i,j} U_{ij} a_i^\dagger a_j^\dagger a_j a_i \tag{3}$$

where $a_i^\dagger$ ($a_i$) create (annihilate) a boson on dimer $i$, and $h = g\mu_B H$ is the effective field. The term $t_{ij}$ defined by the transverse component of the *inter*-dimer interactions $J_1^{x,y}$ etc describes hopping between sites $i$ and $j$ thus endowing the triplons with kinetic energy, see Fig. 1b. $U_{ij}$ from the longitudinal components $J_1^z$ etc is the repulsion energy arising when two triplons occupy neighboring sites $i$ and $j$. The density of triplons is directly controlled by the magnetic field that acts thus as a chemical potential for the bosons [38].

Below $H_{c1}$ the ground state is a quantum-disordered paramagnet formed by the singlet sea (triplon vacuum) and can be approximated by the direct product of singlet states on each dimer, $|\psi\rangle_i = |S, S^z\rangle_i$ with $S = S^z = 0$. Once the spin gap is closed at $H_{c1}$, a Bose condensate is formed. Since the bottom of the triplon band is located at a nonzero wavevector $\mathbf{k}_0 = (\frac{\pi}{a}, \frac{\pi}{a})$ (Fig. 1c), the wavefunction of the condensate varies in space as $\exp(i\mathbf{k}_0 \cdot \mathbf{r})$. In this phase the state of an individual dimer is well approximated by a coherent superposition of the singlet and the $S^z = +1$ triplet: $|\psi\rangle_i = \alpha_i(H)|0,0\rangle_i + \beta_i(H)|1,1\rangle_i$, where the amplitudes $\alpha_i$ and $\beta_i$ depend on the magnetic field $H$ [7, 11, 12].

In the spin language, the condensate corresponds to a spontaneous magnetic order formed by the transverse spin components $\langle S_i^x \rangle$ and $\langle S_i^y \rangle$, violating the remaining rotational $O(2)$ symmetry of the Hamiltonian (1). To make the analogy with the traditional BEC manifest, one can form a $U(1)$ order parameter $\langle S_i^x + iS_i^y \rangle$. The phase corresponding to the angle of the spin in the $XY$ plane is thus the phase of the wavefunction in the boson language. At $H_{c1}$ the paramagnetic phase at low fields makes a transition into a canted antiferromagnet with long-range magnetic order in the plane perpendicular to the field [7] (Figs. 1d and 3a). The staggering of the transverse components of magnetisation reflects a nonzero wavevector of the condensate $\mathbf{k}_0$. The critical properties of the magnet in the vicinity of this phase transition are governed by the quantum critical point (QCP) of the BEC universality class located at $T = 0$ and $H = H_{c1}$. Indeed close to $H_{c1}$ the bosons are extremely diluted, and the effects of the interaction become weak, despite its hard-core nature. Close to the QCP the phase boundary $T_c(H)$ follows [7] a power law $T_c \propto (H - H_{c1})^{\phi}$ with a universal critical exponent $\phi = z/d$, which depends only on the dimensionality ($d$) and dynamical critical exponent $z = 2$ for a quadratic triplon energy band. The upper critical dimension for the QCP is $d_c = 2$.

The BEC QPT has been observed in a growing number of dimer based magnetic insulators, such as $ACuCl_3$ (A=Tl, K, $NH_4$), $BaCuSi_2O_6$, $Cu(NO_3)_2{\cdot}2.5D_2O$, $Cs_3Cr_2Br_9$, $(CH_3)_2CHNH_3CuCl_3$, and $(C_4H_{12}N_2)Cu_2Cl_6$ [8, 14, 32, 33, 36, 39, 40, 41, 42, 43, 44, 45, 46, 47]. The physics of triplon condensation presented for the dimer system remains essentially unchanged since these QCPs are in the same universality class. Quasi-one-dimensional arrangements of $S = 1$ moments in spin chains, e.g. Haldane chains, and even field-saturated frustrated antiferromagnets can be described within the same framework, as was successfully done for nickel-based materials and $Cs_2CuCl_4$, respectively [48, 49, 50, 51, 52].

Experimentally, the static and dynamic magnetic properties have been studied at and around the BEC QCPs in such materials by many experimental techniques. The evolution of magnetic excitations across the QCP at $H_{c1}$ in a three-dimensional network of dimers was studied in $TlCuCl_3$, see Fig. 1a, 2a, and 2c [11, 12, 14, 22]. The softening of the triplet mode $S^z = +1$ in the quantum disordered phase, Fig. 2a, is followed by a dramatic change in the nature of the excitation spectrum above the critical field $H_{c1}$. In particular, as can be expected in a system with a spontaneously broken XY symmetry in the plane perpendicular to the applied magnetic field, a 'Goldstone' mode, with a dispersion linear with momentum appears [11, 12].

Historically, the temperature dependence of the magnetisation $m_z(T)$ at fixed field $H > H_{c1}$ in $TlCuCl_3$ (Fig. 2d) and the scaling of the critical temperature $T_c(H)$ were the key experimental

observations that confirmed the theoretical model [7, 8] of the magnon BEC in dimerized quantum antiferromagnets. The minimum and cusp in $m_z(T)$ at the finite-temperature transition can not be explained within simple mean-field theory [53], but are a consequence of magnon condensation [7, 8] and as such also occur in lower-dimensional magnets. For example comprehensive data from bulk experimental measurements establish the phase diagram and critical exponents for $BaCuSi_2O_6$ (Fig. 2b). The temperature crossover exponent is found to be $\phi = 2/3$, as expected for a BEC QCP in three dimensions. However, below 1 K and down to 35 mK the phase boundary unexpectedly becomes linear, indicating that $\phi = 1$ [33, 34, 35, 36]. We discuss this anomaly below. Even in the case where a full condensate does not exist, such as in one-dimensional systems, similar minima in the magnetisation can occur, but now as a simple crossover [54].

## COMPARISON WITH OTHER BOSON SYSTEMS

The correspondence between a Bose gas and an antiferromagnet is summarized in Table I. Despite many similarities, there are also a few important differences between triplons in a magnet and atoms in a Bose gas [17, 18, 19, 20]. Most importantly, the number of atoms is usually controlled directly (microcanonical ensemble), while the number of triplons is typically set by the magnetic field acting like a chemical potential (canonical ensemble).

Considerable differences also exist at the practical level. Triplons are much lighter and have a much higher density than atomic gases. As a result, condensates survive to much higher temperatures: kelvins in magnets, even room temperature in some cases [15], as opposed to nanokelvins in atomic gases. As solid-state systems, they also allow for a variety of static and dynamic probes (magnetisation, specific heat, NMR, neutrons, etc.). On the other hand, the cold atomic systems allow potentially to study some out-of-equilibrium situations that are hard to maintain in a solid-state device since the condensate is still strongly coupled to the dissipative environment.

From the point of view of realization, the cold atomic systems have a high degree of control and tunability in terms of the interactions and structure. Indeed the structure of the lattice and the hopping of the bosons can directly be controlled by varying the strength of an optical lattice. The interaction can also be controlled via a Feshbach resonance, allowing in principle to realize one's pet Hamiltonian. In spin systems the parameters can be changed only in a limited way by the application of pressure or by changing the chemical composition. Efforts in quantum chemistry produced good realizations of one-, two-, and three-dimensional dimer materials, e.g. $TlCuCl_3$, $BaCuSi_2O_6$, and $(C_5H_{12}N)_2CuBr_4$, respectively. In addition, by the very nature of the mapping

from spins to bosons, the spin systems offer a definite advantage in reaching the limit of strong on-site repulsion as well as the effects of interactions between nearest neighbors (see the next section). The spin systems are therefore an optimal starting point to study situations for which these ingredients are important.

The cold atomic systems have the important advantage that the phase $U(1)$ symmetry is exact. In the magnets, the corresponding $O(2)$ symmetry in the plane perpendicular to the magnetic field can be broken by weak anisotropic interactions (dipolar interactions, crystalline anisotropies, spin-orbit coupling). Even when symmetry-breaking terms are weak, they become important at low temperatures and modify the physics in the vicinity of the QCP. The physics of BEC is also altered in the presence of coupling between spins and lattice distortions. Fortunately, it is possible to use our good control and knowledge of the pure BEC system to treat theoretically these weak deviations as well [26, 28, 29, 55, 56]. Finally the spin systems are excellent to study the critical behavior around the quantum critical points, since the boson density can be finely controlled by the external magnetic field, and the system is fully homogeneous. Intrinsic density inhomogeneities induced by the confining trap in cold atomic systems, make a similar study more difficult.

TABLE I: Correspondence between a Bose gas and a quantum antiferromagnet.

| Bose gas | Antiferromagnet |
|---|---|
| Particles | Spin excitations ($S = 1$ quasi-particles) |
| Boson number $N$ | Spin component $S^z$ |
| Charge conservation U(1) | Rotational invariance O(2) |
| Condensate wavefunction $\langle\psi(\mathbf{r})\rangle$ | Transverse magnetic order $\langle S_i^x + iS_i^y\rangle$ |
| Chemical potential $\mu$ | Magnetic field $H$ |
| Superfluid density $\rho_s$ | Transverse spin stiffness |
| Mott insulating state | magnetisation plateau |

## BEYOND SIMPLE BEC

The interplay of an atomic lattice and strong boson interactions leads to new phenomena beyond the simple BEC paradigm. When the boson density approaches a value commensurate with the lattice periodicity, one often finds magnetisation plateaus where the triplon density stays constant in a finite range of magnetic fields at $T = 0$. We already discussed two simple examples of such

plateaus for $H < H_{c1}$ with zero magnetisation and for $H > H_{c2}$ where each site is occupied by one triplon (Fig. 1d). These simple $m_z = 0$ or 1 plateaus correspond to states with an integer number of bosons per unit cell of the lattice. More generally, since in spin systems the $S^z_{mi}S^z_{nj}$ terms translate into triplon repulsion, magnetisation plateaus at fractional fillings can also occur [57, 58]. Such intermediate plateaus arise for strong enough repulsion between spin excitations on adjacent lattice sites and beyond, and correspond in boson language to Mott-insulator phases [10, 59, 60]. In the simplest case (square lattice, repulsive nearest-neighbor interactions) triplons may form an incompressible state by 'crystallizing' in a checkerboard-like pattern (Fig. 3c). Unlike the ground state of an integer magnetisation plateau (Fig. 3b), such a state violates the translational symmetry of the lattice, and the spontaneous order is characterized by an discrete order parameter ($Z_2$ in the case of the square lattice). Fractional plateaus require strong magnon interactions (in comparison to the kinetic energy) and are therefore less robust then integer ones. Nonetheless, plateaus at magnon fillings 1/3, 1/4, 1/8, and recently for other fractions have been observed in $SrCu_2(BO_3)_2$ [61, 62, 63]. In this material the geometrical frustration suppresses the kinetic energy of magnons thus making magnon repulsion more pronounced [64].

Outside the plateaus, magnons can be freely added to the condensate ensuring a continuously varying magnetisation along the applied magnetic field. It has transverse magnetic order violating the continuous $O(2)$ symmetry, is therefore gapless, and corresponds to the superfluid phase of bosons. Its excitations are Goldstone modes, characteristics of this superfluid phase [11, 14]. The incompressible plateau state and the gapless state with a continuously varying magnetisation are again separated by a QCP.

While the dimensionality of an antiferromagnet is determined mostly by quantum chemistry, it can also change on the fly as the magnet is cooled down. The "Han purple" $BaCuSi_2O_6$ contains layers of dimers with substantial exchange couplings between the layers. Yet the critical behavior exhibits a surprising crossover from three-dimensional at high temperatures to two-dimensional close to the QCP [33, 36]. Two effects contribute to this dimensional reduction at low temperatures. The interplane triplon hopping is suppressed by geometrical frustration: it vanishes exactly at the wavevector of the condensate [33, 34]. Additionally, there are inequivalent layers with different values of the spin gap [31, 35, 36].

Another spectacular example of reduced dimensionality is provided by $(C_5H_{12}N)_2CuBr_4$ [65, 66] and related organo–metallic compounds. These materials can be thought of as a collection of chains of dimers nearly decoupled from each other. As a result, a large part of the $H-T$ phase diagram can be understood by starting in the one-dimensional limit [7, 67, 68, 69, 70, 71]. At low energies

the triplons on a single chain form a Luttinger liquid [72]. Such a compound is thus particularly suited to address the interesting question of the dimensional crossover between a one– and three–dimensional system. In the former, the spins will more behave as fermions, as is well known from the Jordan-Wigner transformation, and in the latter, at temperatures and fields for which residual exchange coupling between the ladders becomes important, the spins are much more behaving as "regular bosons" [73]. The excellent degree of control over the density by the magnetic field as well as the various probes providing access to the dynamical spin-spin correlations make quasi-one-dimensional magnets ideal for studying the dimensional crossover, a phenomenon of general importance for several major areas in physics [72].

We would like to mention the tantalizing possibility of finding an exotic phase known as the supersolid [74]. A hybrid of a solid and a superfluid, this phase violates both the translational symmetry by forming a density wave and the $U(1)$ phase symmetry by exhibiting transverse magnetic order (Figs. 3b-c). It has been conjectured long ago [75, 76, 77] and its existence is under intense debate at the moment following experiments in helium [78, 79]. Such a phase could occur at the end of a fractional plateau because the transition must accomplish two tasks: create a superfluid and destroy a solid (i.e. restore the broken translational symmetry). According to Landau's theory of phase transitions, such transitions are either discontinuous or happen in two stages. Several theoretical calculations show that supersolids exist in realistic magnetic models [80, 81, 82, 83, 84, 85]. The observation of such a phase would thus be an interesting realization of the physics of interacting bosons on a lattice.

Last but not least, advances in growth techniques may allow in the future to study the effects of disorder on bosons [60, 86, 87] and the Bose glass by creating bond disorder, and the influence of impurities on the quantum-critical states [88, 89]. Other directions include the thermodynamics of strongly interacting bosons [37, 90] and quantum coherence at the mesoscopic scale [91].

## CONCLUSIONS AND OUTLOOK

Quantum spins and quantum dimer systems offer remarkable opportunities to study phenomena related to the Bose-Einstein condensation of interacting quantum particles. The high density and low mass of spin excitations, magnons or triplons, leads to a robust nature of the condensate, which survives to temperatures as high as 10 K. The availability of numerous model magnets and physical probes has enabled a detailed study of the critical phenomena and magnetic properties in the vicinity of the BEC quantum phase transition. The spin systems have thus proven to be nicely

complementary to other systems such as the cold atomic gases for the investigation of BEC.

In addition to providing a direct connection with the remarkable BEC phenomenon, the boson picture offers an intuitive understanding for complex quantum properties of matter that are much less physically transparent in the original spin language, and provides access to a cornucopia of exciting new problems and questions. For example, a classical description as canted antiferromagnets of the dimerized magnetic materials discussed here in the field-induced order phase misses up to 100% of the relevant spin physics close to the intrinsic quantum phase transition. Here the possibility to fine tune the density of bosons by using the magnetic field provides a tool to determine the role of the interactions in such systems and a wide range of lattice geometries and dimensionalities is available for study. In combination with the large number of experimental probes for spins this allows to challenge our knowledge of exotic phases of strongly interacting quantum particles, such as BEC in various dimensions, Luttinger-liquid physics, commensurate solids with a fractional number of bosons per unit cell, and supersolids combining superfluidity with a broken translational symmetry.

**Acknowledgments**

This work was supported in part by the Swiss NSF under NCCR MaNEP, a Wolfson Royal Society Research Merit Award, and the US National Science Foundation.

**Competing financial interests**

The authors declare no competing financial insterests.

Correspondence to T.G., thierry.giamarchi@physics.unige.ch, C.R., c.ruegg@ucl.ac.uk, or O.T., olegt@jhu.edu.

[1] Bose, S. N. Plancks Gesetz und Lichtquantenhypothese. *Z. Phys.* **26**, 178 (1924).

[2] Einstein, A. Quantentheorie des einatomigen idealen Gases. *Sitz. Ber. Kgl. Preuss. Akad. Wiss.*, 261 (1924).

[3] Bloch, F. Zur Theorie des Ferromagnetismus. *Z. Physik* **61**, 206 (1930).

[4] Matsubara, T. & Matsuda, H. A lattice model of liquid helium. *Prog. Theor. Phys.* **16**, 569 (1956).

[5] Batyev, E. G. & Braginskii, L. S. Antiferromagnet in a strong magnetic field: analogy with Bose gas. *Sov. Phys. JETP* **60**, 781 (1984).

[6] Affleck, I. Theory of Haldane-gap antiferromagnets in applied fields. *Phys. Rev. B* **41**, 6697 (1990).

[7] Giamarchi, T. & Tsvelik, A. M. Coupled ladders in a magnetic field. *Phys. Rev. B* **59**, 11398 (1999).

[8] Nikuni, T., Oshikawa, M., Oosawa, A. & Tanaka, H. Bose-Einstein condensation of diluted magnons in $TlCuCl_3$. *Phys. Rev. Lett.* **84**, 5868 (2000).

[9] Wessel, S., Olshanii, M. & Haas, S. Field-induced magnetic order in quantum spin liquids. *Phys. Rev. Lett.* **87**, 206407 (2001).

[10] Rice, T.M. To condense or not to condense. *Science* **298**, 760 (2002).

[11] Matsumoto, M., Normand, B., Rice, T. M. & Sigrist, M. Magnon dispersion in the field-induced magnetically ordered phase of $TlCuCl_3$. *Phys. Rev. Lett.* **89**, 077203 (2002).

[12] Matsumoto, M., Normand, B., Rice, T. M. & Sigrist, M. Field- and pressure-induced magnetic quantum phase transitions in $TlCuCl_3$. *Phys. Rev. B* **69**, 054423 (2004).

[13] Nohadani, O., Wessel, S., Normand, B. & Haas, S. Universal scaling at field-induced magnetic phase transitions. *Phys. Rev. B* **69**, 220402 (2004).

[14] Rüegg, C. *et al.* Bose-Einstein condensation of the triplet states in the magnetic insulator $TlCuCl_3$. *Nature* **423**, 62 (2003).

[15] Demokritov, S.O. *et al.* Bose-Einstein condensation of quasi-equilibrium magnons at room temperature under pumping. *Nature* **443**, 430 (2006).

[16] Kasprzak, J. *et al.* Bose-Einstein condensation of exciton polaritons. *Nature* **443**, 409 (2006).

[17] Pitaevskii, L. & Stringari, S. *Bose-Einstein Condensation* (Clarendon Press, Oxford, 2003).

[18] Greiner, M., Mandel, O., Esslinger, T., Hänsch, T. W. & Bloch, I. Quantum phase transition from a superfluid to a Mott insulator in a gas of ultracold atoms. *Nature* **415**, 39 (2002).

[19] Bloch, I. Ultracold quantum gases in optical lattices. *Nature Physics* **1**, 23 (2005).

[20] Bloch, I., Dalibard, J. & Zwerger, W. Many-body physics with ultracold gases. *Rev. Mod. Phys. (in press) and arXiv:cond-mat/0704.3011.*

[21] Cavadini, N. *et al.* Magnetic excitations in the quantum spin system $TlCuCl_3$. *Phys. Rev. B* **63**, 172414 (2001).

[22] Cavadini, N. *et al.* Triplet excitations in low-$H_c$ spin gap systems $KCuCl_3$ and $TlCuCl_3$: an inelastic neutron scattering study. *Phys. Rev. B* **65**, 132415 (2002).

[23] Johannsen, N., Vasiliev, A., Oosawa, A., Tanaka, H. & Lorenz, T. Magnetoelastic coupling in the spin-dimer system $TlCuCl_3$. *Phys. Rev. Lett.* **95**, 017205 (2005).

[24] Vyaselev, O., Takigawa, M., Vasiliev, A., Oosawa, A. & Tanaka, H. Field-induced magnetic order and simultaneous lattice deformation in $TlCuCl_3$. *Phys. Rev. Lett.* **92**, 207202 (2004).

[25] Sherman, E. Y., Lemmens, P., Busse, B., Oosawa, A. & Tanaka, H. Sound attenuation study on the Bose-Einstein condensation of magnons in $TlCuCl_3$. *Phys. Rev. Lett.* **91**, 057201 (2003).

[26] Misguich, G. & Oshikawa, M. Bose-Einstein condensation of magnons in $TlCuCl_3$: Phase diagram and specific heat from a self-consistent Hartree-Fock calculation with a realistic dispersion relation. *J. Phys. Soc. Jpn.* **73**, 3429 (2004).

[27] Glazkov, V. N., Smirnov, A. I., Tanaka, H. & Oosawa, A. Spin-resonance modes of the spin-gap magnet

$TlCuCl_3$. *Phys. Rev. B* **69**, 184410 (2004).

[28] Kolezhuk, A. K., Glazkov, V. N., Tanaka, H. & Oosawa, A. Dynamics of an anisotropic spin dimer system in a strong magnetic field. *Phys. Rev. B* **70**, 020403 (2004).

[29] Sirker, J., Weisse, A. & Sushkov, O.P. Consequences of spin-orbit coupling for the Bose-Einstein condensation of magnons. *Europhys. Lett.* **68**, 275 (2004).

[30] Sasago, Y., Uchinokura, K., Zheludev, A. & Shirane, G. Temperature-dependent spin gap and singlet ground state in $BaCuSi_2O_6$. *Phys. Rev. B* **55**, 8357 (1997).

[31] Rüegg, C. *et al.* Multiple magnon modes and consequences for the Bose-Einstein condensed phase in $BaCuSi_2O_6$. *Phys. Rev. Lett.* **98**, 017202 (2007).

[32] Jaime, M. *et al.* Magnetic-field-induced condensation of triplons in Han Purple pigment $BaCuSi_2O_6$. *Phys. Rev. Lett.* **93**, 087203 (2004).

[33] Sebastian, S.E. *et al.* Dimensional reduction at a quantum critical point. *Nature* **441**, 617 (2006).

[34] Batista, C.D. *et al.* Geometric frustration and dimensional reduction at a quantum critical point. *Phys. Rev. Lett.* **98**, 257201 (2007).

[35] Rösch, O. & Vojta, M. Reduced dimensionality in layered quantum dimer magnets: frustration vs. inhomogeneous condensates. *arXiv:0705.1351*.

[36] Krämer, S. *et al.* Nuclear magnetic resonance evidence for a strong modulation of the Bose-Einstein condensate in $BaCuSi_2O_6$. *Phys. Rev. B* **76**, 100406 (2007).

[37] Xu, G. Y., Broholm, C., Reich, D.H. & Adams, M.A. Triplet waves in a quantum spin liquid. *Phys. Rev. Lett.* **84**, 4465 (2000).

[38] Some purists object to the use of "chemical potential" for the magnetic field term. Note that here the total component of $S^z$ commutes with the Hamiltonian so that the total magnetisation along $z$ and thus the total number of bosons is conserved. We also refer those purists to Shakespeare, Romeo and Juliet II.2.43: "What's in a name? That which we call a rose By any other word would smell as sweet". We thus stick to the terminology chemical potential in the following.

[39] Shiramura, W. *et al.* magnetisation plateaus in $NH_4CuCl_3$. *J. Phys. Soc. Jpn.* **67**, 1548 (1998).

[40] Rüegg, C. *et al.* Neutron scattering study of the field-dependent ground state and the spin dynamics in spin-one-half $NH_4CuCl_3$. *Phys. Rev. Lett.* **93**, 037207 (2004).

[41] Matsumoto, M. Microscopic model for the magnetisation plateaus in $NH_4CuCl_3$. *Phys. Rev. B* **68**, 180403(R) (2003).

[42] Grenier, B. *et al.* Spin excitations throughout the field-induced magnetic phase of the spin 1/2 alternating chain compound $Cu(NO_3)_2 \cdot 2.5D_2O$. *J. Mag. Mag. Matter* **310**, 1269 (2007).

[43] Grenier, B. *et al.* Ordering and excitations in the field-induced magnetic phase of $Cs_3Cr_2Br_9$. *Phys. Rev. Lett.* **92**, 177202 (2004).

[44] Garlea, V. O. *et al.* Excitations from a Bose-Einstein condensate of magnons in coupled spin ladders. *Phys. Rev. Lett.* **98**, 167202 (2007).

[45] Masuda, T. *et al.* Dynamics of composite Haldane spin chains in IPA-$CuCl_3$. *Phys. Rev. Lett.* **96**,

047210 (2006).

[46] Stone, M. B. *et al.* Quantum criticality in an organic magnet. *Phys. Rev. Lett.* **96**, 257203 (2006).

[47] Stone, M. B. *et al.* Field-driven phase transitions in a quasi-two-dimensional quantum antiferromagnet. *New J. Phys.* **9**, 31 (2007).

[48] Zheludev, A. *et al.* Quasielastic neutron scattering in the high-field phase of a Haldane antiferromagnet. *Phys. Rev. Lett.* **88**, 077206 (2002).

[49] Zapf, V.S. *et al.* Bose-Einstein condensation of S=1 nickel spin degrees of freedom in $NiCl_2$-$4SC(NH_2)_2$. *Phys. Rev. Lett.* **96**, 077204 (2006).

[50] Zvyagin, S.A. *et al.* Magnetic excitations in the spin-1 anisotropic Heisenberg antiferromagnetic chain system $NiCl_2$-$4SC(NH_2)_2$. *Phys. Rev. Lett.* **98**, 047205 (2007).

[51] Coldea, R. *et al.* Direct measurement of the spin Hamiltonian and observation of condensation of magnons in the 2D frustrated quantum magnet $Cs_2CuCl_4$. *Phys. Rev. Lett.* **88**, 137203 (2002).

[52] Radu, T. *et al.* Bose-Einstein condensation of magnons in $Cs_2CuCl_4$. *Phys. Rev. Lett.* **95**, 127202 (2005).

[53] Tachiki, M. & Yamada, T. Spin ordering in a spin-pair system. *J. Phys. Soc. Jpn.* **28**, 1413 (1970).

[54] Maeda, Y., Hotta, C. & Oshikawa, M. Universal temperature dependence of the magnetisation of gapped spin chains. *Phys. Rev. Lett.* **99**, 057205 (2007).

[55] Matsumoto, M. & Sigrist, M. Ehrenfest relations and magnetoelastic effects in field-induced ordered phases. *J. Phys. Soc. Jpn.* **74**, 2310 (2005).

[56] Orignac, E. & Citro, R. Magnetostriction in an array of spin chains under a magnetic field. *Phys. Rev. B* **71**, 214419 (2005).

[57] Oshikawa, M., Yamanaka, M. & Affleck, I. magnetisation plateaus in spin chains: "Haldane gap" for half-integer spins. *Phys. Rev. Lett.* **78**, 1984 (1997).

[58] Cabra, D. C., Honecker, A. & Pujol, P. magnetisation plateaux in N-leg spin ladders. *Phys. Rev. B* **58**, 6241 (1998).

[59] Haldane, F. D. M. Effective harmonic-fluid approach to low-energy properties of one-dimensional quantum fluids. *Phys. Rev. Lett.* **47**, 1840 (1981).

[60] Fisher, M. P. A., Weichman, P. B., Grinstein, G. & Fisher, D. S. Boson localization and the superfluid-insulator transition. *Phys. Rev. B* **40**, 546 (1989).

[61] Kageyama, H. *et al.* Exact dimer ground state and quantized magnetisation plateaus in the two-dimemsional spin system $SrCu_2(BO_3)_2$. *Phys. Rev. Lett.* **82**, 3168 (1999).

[62] Kodama, K. *et al.* Magnetic superstructure in the two-dimensional quantum antiferromagnet $SrCu_2(BO_3)_2$. *Science* **298**, 395 (2002).

[63] Sebastian, S.E. *et al.* Fractalization drives crystalline states in a frustrated spin system. *arXiv:0707.2075*.

[64] Miyahara, S. & Ueda, K. Theory of the orthogonal dimer Heisenberg spin model for $SrCu_2(BO_3)_2$. *J. Phys.: Condens. Matter* **15**, R327 (2003).

[65] Watson, B.C. *et al.* Magnetic spin ladder $(C_5H_{12}N)_2CuBr_4$: High-field magnetisation and scaling near quantum criticality. *Phys. Rev. Lett.* **86**, 5168 (2001).

[66] Lorenz, T. *et al.* Diverging thermal expansion of the spin-ladder system $(C_5H_{12}N)_2CuBr_4$. *arXiv:0711.1510*.

[67] Chitra, R. & Giamarchi, T. Critical properties of gapped spin-1/2 chains and ladders in a magnetic field. *Phys. Rev. B* **55**, 5816 (1997).

[68] Mila, F. Ladders in a magnetic field: a strong coupling approach. *Eur. Phys. J. B* **6**, 201 (1998).

[69] Furusaki, A. & Zhang S.-C. Dynamical spin correlations in the Heisenberg ladder under a magnetic field and correlation functions in the SO(5) ladder. *Phys. Rev. B* **60**, 1175 (1999).

[70] Usami, M. & Suga, S. Critical properties of S=1/2 Heisenberg ladders in magnetic fields. *Phys. Rev. B* **58**, 14401 (1998).

[71] Hikihara, T. & Furusaki, A. Spin correlations in the two-leg antiferromagnetic ladder in a magnetic field. *Phys. Rev. B* **63**, 54438 (2001).

[72] Giamarchi, T. *Quantum Physics in One Dimension* (Oxford Univ. Press, Oxford, 2004).

[73] Orignac, E., Citro, R. & Giamarchi, T. Critical properties and Bose-Einstein condensation in dimer spin systems. *Phys. Rev. B* **75**, 140403 (2007).

[74] Prokof'ev, N. & Svistunov, B. Supersolid state of matter. *Phys. Rev. Lett.* **94**, 155302 (2005).

[75] Andreev, A. F. & Lifshitz, I. M. Quantum theory of defects in crystals. *Sov. Phys. JETP* **29**, 1107 (1969).

[76] Chester, G. V. Speculations on Bose-Einstein condensation and quantum crystals. *Phys. Rev. A* **2**, 256 (1970).

[77] Leggett, A. J. Can a solid be superfluid. *Phys. Rev. Lett.* **25**, 1543 (1970).

[78] Kim, E. & Chan, M. H. W. Probable observation of a supersolid helium phase. *Nature* **427**, 225 (2004).

[79] Sasaki, S., Ishiguro, R., Caupin, F., Maris, H. J. & Balibar, S. Superfluidity of grain boundaries and supersolid behavior. *Science* **313**, 1098 (2006).

[80] Wessel, S. & Troyer, M. Supersolid hard-core bosons on the triangular lattice. *Phys. Rev. Lett.* **95**, 127205 (2005).

[81] Heidarian, D. & Damle, K. Persistent supersolid phase of hard-core bosons on the triangular lattice. *Phys. Rev. Lett.* **95**, 127206 (2005).

[82] Melko, R. G. *et al.* Supersolid order from disorder: Hard-core bosons on the triangular lattice. *Phys. Rev. Lett.* **95**, 127207 (2005).

[83] Ng, K. K. & Lee, T. K. Supersolid phase in spin dimer XXZ systems under a magnetic field. *Phys. Rev. Lett.* **97**, 127204 (2006).

[84] Sengupta, P. & Batista, C.D. Field-induced supersolid phase in spin-one Heisenberg models. *Phys. Rev. Lett.* **98**, 227201 (2007).

[85] Laflorencie, N. & Mila, F. Quantum and thermal transitions out of the supersolid phase of a 2D quantum antiferromagnet. *Phys. Rev. Lett.* **99**, 027202 (2007).

[86] Giamarchi, T. & Schulz, H. J. Anderson localization and interactions in one-dimensional metals. *Phys. Rev. B* **37**, 325 (1988).

[87] Nohadani, O., Wessel, S. & Haas, S. Bose-glass phases in disordered quantum magnets. *Phys. Rev. Lett.* **95**, 227201 (2005).

[88] Mikeska, H.-J., Ghosh, A. & Kolezhuk, A. K. Field-induced disorder in a gapped spin system with nonmagnetic impurities. *Phys. Rev. Lett.* **93**, 217204 (2004).

[89] Xu, G. *et al.* Holes in a quantum spin liquid. *Science* **289**, 419 (2000).

[90] Rüegg, C. *et al.* Quantum statistics of interacting dimer spin systems. *Phys. Rev. Lett.* **95**, 267201 (2005).

[91] Xu, G. *et al.* Mesoscopic phase coherence in a quantum spin fluid. *Science* **317**, 1049 (2007).

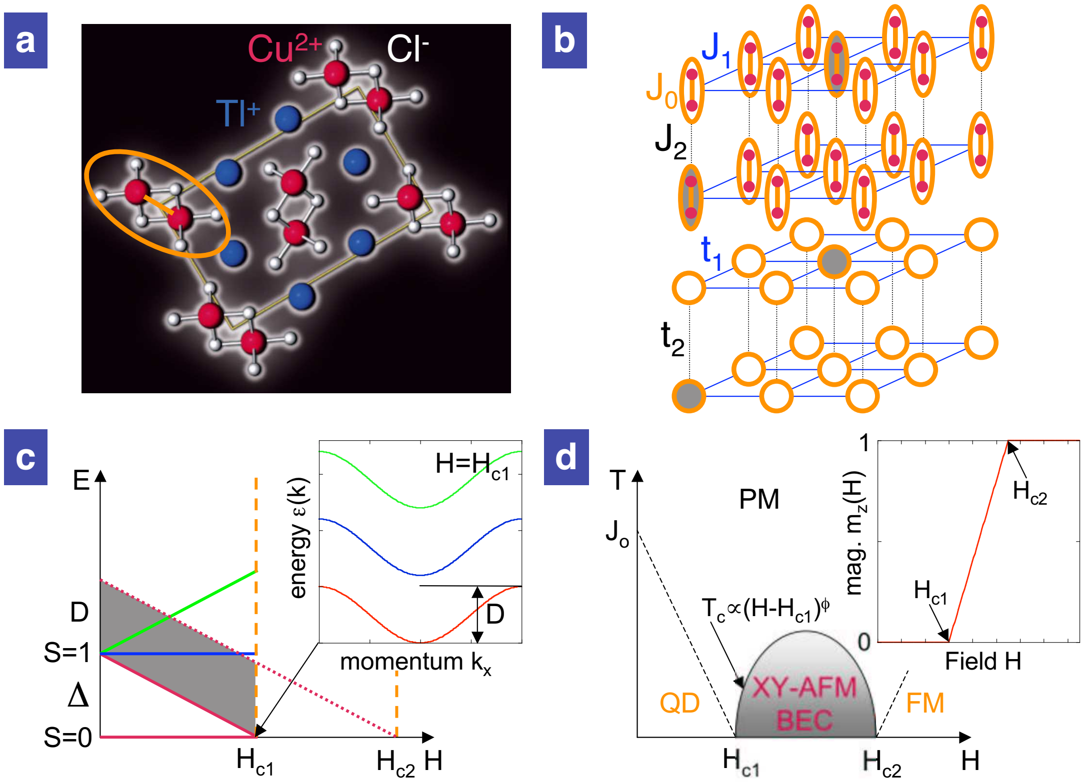


FIG. 1: BEC of magnons in dimerized quantum antiferromagnets: (a) Dimers in the real material $TlCuCl_3$ with $S = 1/2$ from $Cu^{2+}$ ions and superexchange via $Cl^-$ [8, 11, 12, 14, 21, 22, 90]. (b) Dimers on a square lattice with dominant antiferromagnetic intradimer interaction $J_0$ and interdimer interactions $J_i$. Triplet states (gray, top) are mapped on quasi-particle bosons (triplons, bottom). (c) Zeeman splitting of the triplet modes with gap $\Delta$ and bandwidth $D$ at $\mathbf{k}_0 = (\frac{\pi}{a}, \frac{\pi}{a})$. Dispersion of triplons at the critical field $H_{c1}$ [11, 12, 14, 21, 22]. (d) Resulting phase diagram with paramagnetic (PM), quantum disordered (QD), field-aligned ferromagnetic (FM), and canted-antiferromagnetic (XY-AFM) phase, where BEC of magnons occurs. Close to $H_{c1}$ and $H_{c2}$ the phase boundary follows a power–law $T_c \propto (H - H_{c1})^{\phi}$ with a universal exponent $\phi = 2/3$ for BEC of magnons [7, 8]. Magnetisation curve $m_z(H)$ for 3D dimer spin system with plateau at $m_z = 0, 1$ (gapped) [8, 32, 33].

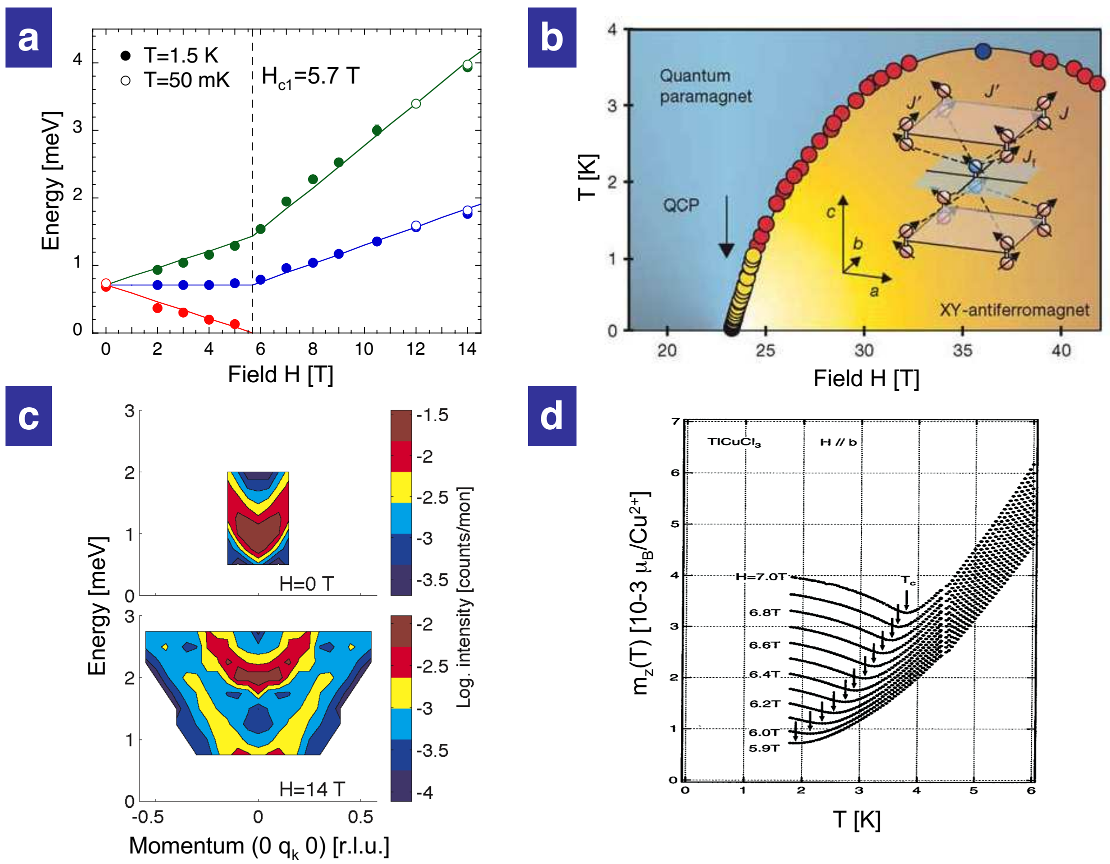


FIG. 2: Experimental results on magnon BEC: (a) Zeeman splitting of the triplet modes in $TlCuCl_3$ up to $H > H_{c1}$ measured by inelastic neutron scattering [14, 27, 28]. (b) Phase diagram of $BaCuSi_2O_6$ measured by torque magnetisation, magneto-caloric effect, and specific heat [32, 33]. Dimensional reduction was reported in this material with a crossover from the 3D-BEC critical exponent $\phi = 2/3$ to $\phi = 1$ for 2D at temperatures close to the QCP [33]. (c) Excitations in the BEC of triplons realized in $TlCuCl_3$ [14, 27, 28]. Goldstone mode with linear dispersion around $\mathbf{k}_0$. Spin anisotropy generally leads to a spin gap in real materials [27, 28, 29]. (d) Temperature-dependence of the magnetisation $m_z(T)$ in $TlCuCl_3$ for fixed magnetic field $H$, as indicated [8]. Minima at the finite-temperature phase transition (vertical arrows), as expected for a BEC of triplons.

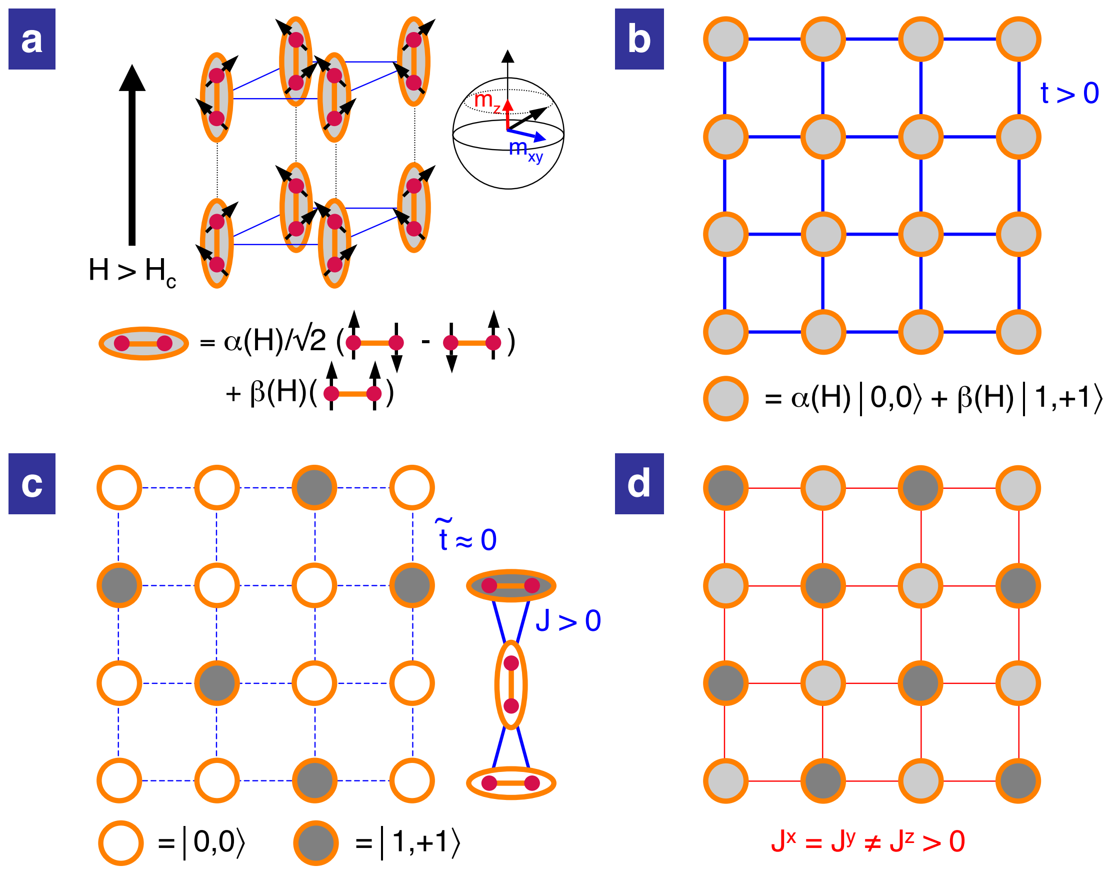


FIG. 3: Lattice boson phases: (a) Canted antiferromagnetic state at $H_{c1} < H < H_{c2}$ in a BEC of magnons. Longitudinal $m_z$, see Fig. 1d, and transverse $(XY)$ magnetisation $m_{xy}$. (b) Triplon condensate in superfluid phase. The hopping amplitude $t$ of the quasi-particles leads to a uniform mixture of singlet and triplet states. (c) Analog of the Mott-insulating phase for triplons with magnetisation plateau at $m_z = 1/3$: triplons "crystallize" to form a superstructure. This phase is partially observed in $SrCu_2(BO_3)_2$, where quasi-particle hopping is strongly suppressed by geometrical frustration [10, 61, 62, 64]. (d) Supersolid on a square-lattice of dimers with exchange anisotropy as proposed theoretically [83, 85], and for other lattice geometries [80, 81, 82, 84]. Both translation and $U(1)$ symmetry are spontaneously broken, i.e. coexistence of superfluid and "crystal" of lattice bosons.